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Original Article

Measurement of radioactivity in spa waters using gamma spectrometry and evaluation of health risks

Raouen Graichi*, Linda Chabouni, Neserine Hamitouche, Ali Benbourenane, Omar Amri

Radiological Expertise Department, Nuclear Research Center of Algiers, 02, Bd Frantz Fanon B.P.399Alger-Gare, Algeria

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ABSTRACT

Exposure of human beings to radiation from natural sources is a permanent and unavoidable part of life on earth, in this study; we are interested in measuring the concentrations of naturally occurring radionuclides in spa water, such as ^{40}K, ^{232}Th, ^{235}U, ^{226}Ra and ^{238}U, using gamma spectrometry with a hyper pure germanium (HPGe) detector. We analysed four samples from different regions of Algeria. The samples were imbedded in 1L vials and stored for at least 21 days to achieve secular equilibrium between ^{226}Ra and its short-lived daughter products before analysing by gamma ray spectrometry. To assess the radiological effects of these four samples, we calculated the annual effective dose AED. The doses received by the public are calculated based on the values of specific activities of ^{226}Ra, ^{232}Th, ^{238}U, ^{40}K and ^{235}U. The average activity concentrations for the samples were 7.4± 0.7 to 10.78± 0.70 Bq/L for ^{238}U; 1.53± 0.09 to 3.5± 0.15 for ^{226}Ra; 0.08± 0.01 to 1.4± 0.08 for ^{232}Th; 2.30± 0.12 to 5.51± 0.2 for ^{235}U and 8.06± 0.4 to 40.30± 2.11 for ^{40}k.The estimated doses for ^{226}Ra exceed the WHO and UNSCEAR recommended values of 0.26 mSv/y and 0.29 mSv/y respectively in all samples. The total annual effective doses for sample S02 exceed the ICRP recommended limit 1 mSv/y.

1. Introduction

The presence of naturally occurring radioactivity in water is a result of the surrounding geological environment. As water travels through springs, streams, lakes, reservoirs, and aquifers, when it crosses rocks and sediments containing radioactive elements such as uranium, thorium, and radium, it has the potential to engage with them. This interaction may lead to the dissolution of these elements in the water, resulting in varying levels of natural radioactivity.

Recently, spa waters have gained popularity among individuals for their perceived health benefits, both for therapeutic use and consumption. Consequently, numerous researchers have undertaken radioactivity assessments in spas located in various regions across the world [1-13].

The World Health Organization (WHO) has provided recommended safe levels for different parameters that define the quality of drinking water in its general guidelines [14]. Many countries have implemented these guidelines to set up their own specific water quality standards at the national level, whereas there are still some that have not developed their standards.

The existence of radioactive materials that can be ingested or inhaled may result in adverse biological effects and represent risks to human health. To clarify, when radionuclides and heavy metals are found in water sources, they can result in internal exposure as these radionuclides decay following absorption by the human body [15].

Precisely quantifying the activity concentration of naturally occurring radionuclides in drinking water is essential for assessing the extent of ionizing radiation exposure to the

* *Corresponding author address:*
E-mail address: r.graichi@crna.dz

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human population through ingestion and domestic use. This is critical because the radiation doses from these routes are directly linked to the quantity of radionuclides present, it serves as a significant factor in ensuring radiological safety for the population concerning drinking water [15–18], tap water [19], as well as stream or surface water [20–23]. The most prevalent Radionuclides detected in water are ^{238}U, ^{226}Ra, ^{235}U, ^{232}Th and ^{40}K. The radionuclides that pose the highest radio toxicity and danger include radium, which exhibits similar behavior to calcium once it is absorbed into the body. Prolonged internal exposure of humans to elevated radium levels can lead to the development of bone and sinus cancers. The objective of this study is to assess the gamma activity concentrations of natural isotopes from the ^{238}U series, ^{232}Th series, and ^{40}K using an HPGe detector system. Furthermore, we estimate the annual effective doses associated with the consumption of these waters based on the concentration values. These measurements were conducted at the Nuclear Research Center of Algiers (CRNA), Algeria. conducted at the Nuclear Research Center of Algiers (CRNA), Algeria.

2. Materials and Methods

2.1. Sample collection methodology

We collected four water samples from various locations from December to January of the year 2022-2023.The specific locations are indicated in Fig1, and their coordinates in Table 1.

Fig 1. Map showing samples locations.

Table 1: Coordinates of the geographical locations of the analyzed thermal springs.

Sample code	Thermal water name	Province	Geographic coordinate
S01	El Biban	Borj Bou Arreridj	36°11'47"N 4°23'20" E
S02	Guergour	Setif (bougaa)	36° 19' 00" N 5° 04' 00" E
S03	Malouane	Blida	36°29'12.7"N 3°02'37.8"E
S04	Teleghma	Mila	36° 06′ 55″ N, 6° 21′ 51″ E

Samples were obtained directly from the thermal source and placed in 1.5 Liter plastic bottles using established closed-source techniques. These bottles were properly labeled, indicating both the date and location of collection. Each water sample was transferred into a sealed 1liter vial and stored for a minimum of 21 days before being subjected to gamma spectrometry analysis. This waiting period allows the daughter products to reach a state of secular equilibrium with their corresponding radionuclide parent [24].

2.2. Analytical methods of radioactivity

The activity concentrations in the water sample were measured with high-purity (HPGe) open co-axial detectors, specifically utilizing the Canberra GC 3018 model bearing the serial number B15079, which was connected to a Canberra Multichannel Analyzer (MCA) computer system. The Energy calibrations of the Spectrometer were performed by utilizing gamma-ray sources emitting in the energy range of 59.5-1332.5 keV, including ^{241}Am, ^{137}Cs, and ^{60}Co. The energy calibration curve is depicted in Figure 2.

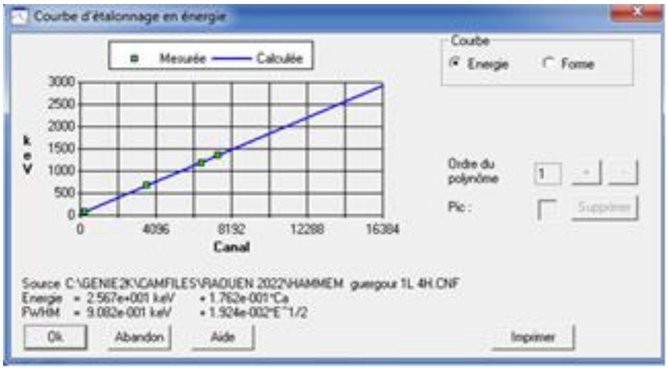

Fig 2. Energy calibration curve.

The Efficiency calibrations for the spectrometer were conducted using standard ^{152}Eu source (water contaminated with radioactive source of ^{152}Eu). The energy spectra encompass a range from 39.52 to 1408.01 KeV, ensuring the coverage of all relevant gamma energies from the radionuclides of interest. Fig 3 shows the efficiency calibration curve.

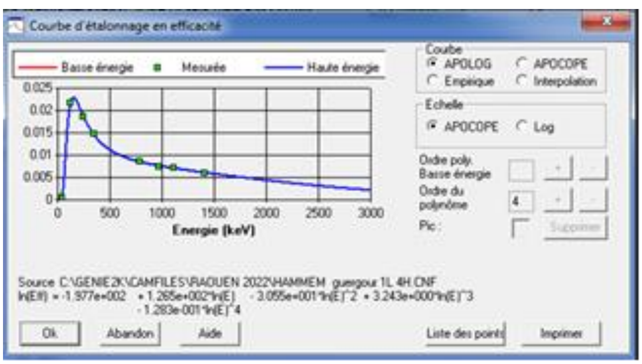

Fig 3. Efficiency calibration.

After equilibrium, each sample was placed on top of the detector and counted for 14400s.the background was measured using a 1L empty vial (same geometry of samples) for the same counting times. The recurring distinctive photo peaks in the sample spectra were attributed to radionuclides from the natural decay chains of ^{238}U, ^{232}Th, and ^{40}K. The determination of the parent radionuclides relies on the energy peaks of gamma rays emitted by the daughter products in equilibrium with their parent nuclides.

The figure below presents a sample spectrum from sample S01, which was acquired using genie 2000 software.

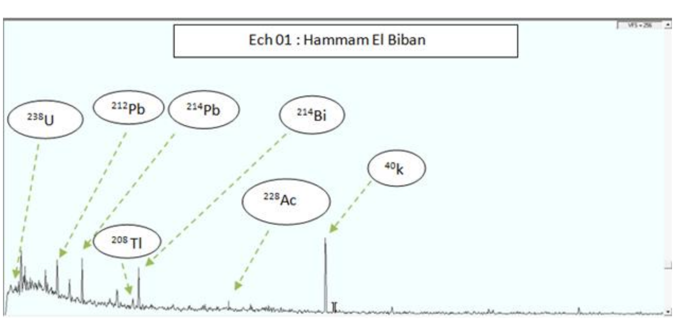

Fig 4. Sample S01 Spectrum (Hammam el Biban)

The radionuclide activities were determined utilizing the following equation:

$$A = \left[\frac{N}{I \times T \times \varepsilon \times P} \right] \quad (1$$

In this equation, "N" represents the net gamma ray counting rate, "ε" denotes the efficiency of the specific gamma ray, "I" signifies the absolute transition probability of gamma decay, "P" stands for the sample's volume, and "T" represents the counting duration.

The assessment of ^{226}Ra and ^{232}Th relies on the identification of gamma-ray energy peaks emitted by the decay products that are in equilibrium with their parent nuclides.

- The activity levels of ^{226}Ra were computed by analyzing the gamma-ray emissions at energies of 609.3 keV (corresponding to ^{214}Bi) and 351.9 keV (corresponding to ^{214}Pb).

- The activity levels of ^{232}Th were determined by examining the gamma-ray emissions at energies of 583.1 keV (corresponding to ^{208}Tl), 283.6 keV (related to ^{212}Pb), and 911.1 keV (related to ^{228}Ac).

- The activity levels of 40K were derived through the detection of the gamma-ray peak at 1460.8 keV.

$$A_{226Ra} = \left[\frac{{}_{214}Bi + {}_{214}Pb}{2} \right] \quad (2$$

$$A_{232Th} = \left[\frac{{}_{208}Tl + {}_{212}Pb + {}_{228}Ac}{3} \right] \quad (3$$

2.3 Calculation of annual effective dose

Spa resorts are experiencing a growing trend in popularity as destinations for therapeutic and relaxation purposes. The annual radiation exposure for individual spa-goers resulting from the presence of elements like ^{226}Ra, ^{232}Th, ^{40}K, and ^{238}U in the spa water is determined using the parameters outlined in the UNSCEAR report of 2000, as described [25]:

$$AED = C \times I \times E \quad (4$$

In this equation, AED represents the yearly effective radiation dose for an individual resulting from the consumption of radionuclides (mSv/y), C stands for the concentration of radionuclides in the ingested water, expressed in Bq per liter (Bq.L–1), I signify the annual intake of drinking water, which is conventionally set at 730 liters annually in accordance with the UNSCEAR's recommendations. and E represents the conversion factor for Internal Radiation Dose caused by the radionuclide (Sv.Bq–1). The specific dose conversion factors employed in this calculation are sourced from the ICRP publication and are detailed in Table 2.

Table 2: dose conversion factors for ingestion of radionclides [26]:

Radioisotope	Dose conversion factors ($Sv.Bq^{-1}$)
^{226}Ra	2.8×10^{-7}
^{235}U	4.7×10^{-8}
^{232}Th	2.3×10^{-7}
^{40}K	6.2×10^{-9}

3. Results and Discussion

Measured activity of ^{238}U, ^{226}Ra, ^{40}K, ^{235}U and ^{232}Th of each sample are presented in Table 3.

The Activities vary from 7.4± 0.7 to 10.78± 0.70 Bq/L for ^{238}U; 1.53± 0.09 to 3.5± 0.15 for ^{226}Ra; 0.08± 0.01 to 1.4± 0.08 for ^{232}Th; 2.30± 0.12 to 5.51± 0.2 for ^{235}U and 8.06± 0.4 to 40.30± 2.11 for ^{40}K.

Table 3: Radionuclides activity in spa water in (Bq/ L).

Radio-nuclides	Energy	Activity (Bq/L)			
		S01	S02	S03	S04
^{214}Bi	609.3	2.00±0.10	2.42±0.01	1.52±0.09	1.54±0.09
^{214}Pb	351.9	3.50±0.15	2.90±0.10	1.80±0.12	1.53±0.09
^{228}Ac	911	0.60±0.07	1.14±0.10	1.13±0.07	1.40±0.08
^{212}Pb	238.6	1.30±0.06	0.90±0.05	1.00±0.05	1.10±0.06
^{208}Tl	583.1	0.40±0.03	0.08±0.01	0.13±0.01	0.30±0.03
^{234}Th	63.3	8.70±0.32	7.40±0.70	<BDL	10.78±0.70
^{235}U	143.7	4.30±0.21	5.51±0.20	<BDL	2.30±0.12
^{40}K	1460.8	40.30±2.11	8.06±0.40	33.62±1.70	11.74±0.63

BDL :below detection limit.

Table 4: Contrasting natural radioactivity levels in water with those in other countries.

country	^{226}Ra	^{232}Th	^{40}K	References
Yemen	3.48 Bq/L	1.01 Bq/L	16.05 Bq/L	[27]
Jordon	3.8 Bq/L	1.42 Bq/L	23.2 Bq/L	[28]
Turkey	BDL-163 mBq/L	BDL-41MBq/L	BDL-511 mBq/L	[29]
Pakistan	1.75 mBq/L	1.34 mBq/L	48.08 mBq/L	[30]
Algeria	2.15 Bq/L	0.79 Bq/L	23.43 Bq/L	This work

The estimated activity concentrations are within the concentration range in Yemen and Jordan and higher than those of Turkey and Pakistan. The observed differences in the specific activity of the radioelements among the four stations are due to local variations in rock formation and the geological characteristics of each area.

The element ^{226}Ra stands out for its exceptional extension of the biological half-life and high water solubility, making it of paramount importance. This element has the potential to introduce contamination into the human body, whether through the consumption of thermal water or through the inhalation of ^{222}Rn during degassing processes occurring within the enclosed environments of various spa facilities.

To assess the radiological risks, we performed an Annual Effective Dose (AED) calculation, and the outcomes of this calculation are presented in Table 5.

Table 5: Annual effective doses in spa water.

Radio-nuclide	AED (mSv/y)			
	S01	S02	S03	S04
^{226}Ra	0.56	0.74	0.33	0.31
^{235}U	0.14	0.18	/	0.07
^{232}Th	0.12	0.11	0.12	0.15
^{40}K	0.18	0.03	0.15	0.05
Cumulative average	1	1.06	0.6	0.58

The yearly effective dose resulting from the ingestion of ^{232}Th ranges from 0.11 to 0.15 mSv/y; from 0.07 to 0.18 mSv/y for ^{235}U; from 0.31 to 0.74 for ^{226}Ra and from 0.03 to 0.18 mSv/y for ^{40}K.

The annual effective doses from ^{226}Ra in this research surpass the recommended reference dose levels of 0.26 mSv/year as set forth by the World Health Organization (WHO) [31] and 0.29 mSv/year as advised by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) [32].

The ICRP guidelines establish a maximum public exposure limit at an effective dose of 1 mSv/year [33-35], The overall annual effective dose falls below the recommended limit of 1 mSv/y set by the International Commission on Radiological Protection (ICRP) for

samples S03 and S04 (Hammam Melouan) and Hammam Tlaghma). It equals 1 mSv/y for sample S01 (Hammam El Biban), while the total annual dose for sample S02 (Hammam Guergour) significantly exceeds the recommended limit.

4. Conclusion

This study aimed to examine the radioactivity levels of ^{226}Ra, ^{232}Th, ^{235}U, ^{238}U, and ^{40}K in spa waters from four regions in Algeria. The significance of these measurements lies in their impact on public health, as these waters are predominantly utilized for therapeutic and drinking purposes without full awareness, due to the widespread belief in their health benefits. The annual radiation exposure doses were computed as part of the analysis. It was observed that the estimated doses for ^{226}Ra surpassed the recommended values of 0.26 mSv/y by WHO and 0.29 mSv/y by UNSCEAR in all samples. Furthermore, the total annual effective doses for sample S02 exceeded the ICRP's recommended limit of 1 mSv/y.

Constantly monitoring radioactivity in spa water is undeniably crucial for radiation protection purposes. The information presented in this study can serve as a reference point for estimating the impact of radioactive pollution in these regions.

Conflict of Interest: The authors declare that they have no conflict of interest.

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